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#### **Final Technical Report**

#### for 30 September 1994 to 29 September 1997

#### submitted by

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## AFOSR-DEPSCoR Contract Number F49620-94-1-0465

#### PERSISTENT SPECTRAL HOLE BURNING MATERIALS FOR TIME- AND FREQUENCY-DOMAIN OPTICAL MEMORIES AND SIGNAL PROCESSING

#### submitted to

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#### **Summary of Objectives and Accomplishments**

Optical memories and optical signal processing based on spectral hole burning (SHB) and optical coherent transients can dramatically increase optical storage density and provide a new range of signalprocessing devices. The search for new optical materials for SHB applications, evaluation of their dynamic and static properties, and studies of the ultimate limits on material performance constitute the research that we carried out under the AFOSR Optoelectronic Information Processing Devices and Systems Program of Dr. Alan E. Craig. We worked to understand the atomic-scale mechanisms that determine material performance, emphasizing parameters relevant to device development. Attention was focused on rare earth and transition metal ion materials; systems for photon-gated SHB were also explored. We designed a range of new crystal materials containing Eu<sup>3+</sup>, Pr<sup>3+</sup>, Tm<sup>3+</sup>, Tb<sup>3+</sup>, and Er<sup>3+</sup> ions and optimized their hole burning and coherent transient properties. Crystal composition and rare earth ion concentration were tailored to specific device needs. Improvements of performance parameters, often by more than an order of magnitude, were achieved for these materials at a variety of wavelengths where diode laser sources make new materials especially important. In the case of Er3+, we made the first reports of any material studied for spectral hole burning and coherent transient applications in the 1.5 µm optical communications bands. Going beyond specific materials, we made a fundamental analysis of crystal symmetry that can guide the choice and design of all future materials.

State of the art facilities for SHB research and SHB material characterization are available in our laboratory, and further development in capabilities occurred during this project. We have eighteen years experience studying a variety of rare earth activated hole burning materials. Our work at Montana State University was carried out in collaboration with *Scientific Materials Corporation — an AFOSR SBIR Phase II contractor*, with Roger Macfarlane at IBM Almaden Research Center, with university groups including Dr. M. J. M. Leask at the University of Oxford, UK, and with other groups in the US and France.

Specific accomplishments included:

- Characterization and optimization of  $Eu^{3+}$ :  $Y_2SiO_5$  for use in prototype optical memory devices (ODRAM), with special consideration of the concentration and temperature dependence of the following optical parameters: absorption coefficient and line shape  $\alpha(\omega)$ , inhomogeneous line width  $\Gamma_{inh}$ , homogeneous linewidth  $\Gamma_h$  and coherence time  $T_2$ , hole lifetimes, spectral diffusion, and instantaneous spectral diffusion.
- 2. Characterization of Pr<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>, with particular attention to instantaneous spectral diffusion effects (laser induced spectral diffusion). This is also relevant to ODRAM.
- Design and characterization of eight  $Tm^{3+}$  compounds, with special consideration of the concentration and temperature dependence of these parameters: absorption coefficient and line shape  $\alpha(\omega)$ , inhomogeneous line width  $\Gamma_{inh}$ , homogeneous linewidth  $\Gamma_h$  and coherence time  $T_2$ , hole lifetimes, spectral diffusion, and instantaneous spectral diffusion.
- 4. Design and characterization of the first Er<sup>3+</sup> materials for devices operating in the 1.5 μm communication bands. Characterization and optimization of eight new Er<sup>3+</sup> materials.
- 5. Design and characterization of Tb<sup>3+</sup> compounds for photon gated SHB, with particular attention to ultraviolet and two-photon spectroscopy of high lying energy levels that are involved in the two-step gating process.

#### **Materials for Specific Applications**

To meet the demand for access and processing of larger and larger amounts of data with ever-increasing speed from remote distances, new devices must be developed. No computer is fast enough to solve the most pressing problems of the moment. The intrinsic quantum-mechanical and optical coherence properties of SHB materials can provide terahertz memory and signal processing speeds. The practical aspects of storage and simultaneous processing in more than 10,000 parallel channels already have been demonstrated by teams at MSU and elsewhere. Exciting features of two applications areas are mentioned as examples:

- 1. High performance memories
  - low latency/high capacity memory
  - high speed recording of massive parallel data streams
  - holographic image recording and analysis
- 2. Ultra-high-speed parallel signal processing
  - optical routers, precision time delays, and time code generators
  - versatile optical frequency standards and high speed optical clocks.

Additional capabilities provided by the technology have led to a broad range of proposed applications including:

All-optical passive router
Frequency multiplexing
Code division multiplexing
Secure communications
Real-time address decoder
Packet header stripper/isolator
FDM optical interconnect
FDM reference filters
Parallel computing
Clocks and Frequency Standards

Associative memory
Look-up tables
Bit rate conversion
Dynamic pulse shaping
Distortion compensation
Dispersion compensation
Target and pattern recognition
True-time delay (w/processing)
Adaptive true-time delay beam forming
Wide-aperture narrow-band filters

Applications that utilize the full multi-dimensional capabilities of coherent spectral hole burning computers are:

- networks with distributed intelligence
- multidimensional and associative/content addressable data retrieval systems;
- analysis and simulation of nervous systems
- optical "brains"

To reach maturity, this technology requires a well-coordinated effort on material development. Material development and characterization were the primary tasks undertaken in this project. Our work was facilitated by close cooperation with SBIR contractor Scientific Materials Corporation of Bozeman, Montana.

#### **Summary of Effort**

Our research on materials for SHB and optical coherent transients included a variety of accomplishments and areas of work. A more complete listing is given here, with detailed reports following in subsequent sections.

a) The interaction mechanisms that establish fundamental limits on material performance were studied. Performance parameters were considered for applications including high-density optical storage and high-speed real-time signal processing.

b) Atomic-scale material properties were determined for Eu<sup>3+</sup>, Pr<sup>3+</sup>, Tm<sup>3+</sup>, and Er<sup>3+</sup> ions in

many hosts.

New materials, specifically for high-density random access optical memories, were designed, evaluated, and developed. For example, a concentration dependence study on Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> compounds was begun in support of a 100 GByte optical dynamic random access memory device (ODRAM) developed for market by DoD SBIR contractors Templex Technologies and Scientific Materials.

New Tm<sup>3+</sup> materials were designed, developed, and characterized for use with diode lasers in the 790 - 800 nm range. Eight Tm<sup>3+</sup> materials were studied in detail. These materials are to be used by a group of DoD investigators in a MURI project for generation of true

time delays in the context of phased array radar.

e) We made the first report of a practical Er<sup>3+</sup> material for SHB applications in the important 1.5 μm communications region. This Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> material enables demonstration of optical routers based on spatial-spectral holography concepts. Demonstration of buffer memories is projected.

f) We devised, developed, and characterized eight new Er<sup>3+</sup> materials for the 1.5 μm communications region, producing materials with varied properties and for a range of

wavelengths.

We made the first report of a practical application of an Er<sup>3+</sup> material in a device based on spatial-spectral holography. An optical header decoder for optical routing successfully operated at 1531 nm. Demonstration of a packet header decoder for all-optical optical packet switching in optical communications, for example along optical fibers: This signal processing was demonstrated in the important 1.5 μm region.

h) These new Er<sup>3+</sup> materials were used by DoD contractor SRI.

i) "Photon gated" spectral hole burning development was carried out. Work focused on investigation of eight different Tb<sup>3+</sup> materials and the search for new ions, new hosts, and new combinations.

provision of solid state frequency references for stable lasers and optical clocks in communications and computer systems using persistent spectral hole burning is another application of interest to our group. Our work on Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> suggests that stability to the milli-Hertz scale should be achievable.

k) Optimization of symmetry considerations in hole burning and coherent transient materials has led us to a general determination of the optimum propagation and light polarization

directions for materials with all crystal symmetries.

We participated in and organized the following AFOSR-sponsored Workshops:

- a) Cone organized the AFOSR-sponsored workshop Material Requirements for Persistent Spectral Hole Burning and Time-Domain Optical Storage and Processing held in Bozeman, MT, on August 3-4, 1994, presented experimental results, and published the proceedings. A number of group members participated.
- b) Cone presented experimental results at the AFOSR Workshop in Seattle on February 1-2, 1995.
- c) Cone assisted with local arrangements for and presented experimental results at the AFOSR Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, March 3-6, 1996. A number of group members participated.
- d) Cone resented experimental results in an invited talk, "Spectral-Hole-Burning Frequency Standards and Stable Lasers," at the AFOSR Contractors Workshop on Time and Frequency Standards, Phillips Laboratory, Kirtland Air Force Base, Albuquerque, NM, April 25, 1996.
- e) Cone presented experimental results at the AFOSR Workshop on Frontiers of Applications of Persistent Spectral Hole Burning, Big Sky, MT, February 16-19, 1997. A number of group members participated.

#### Accomplishments/New Findings

A number of materials were designed jointly with Scientific Materials Corporation and produced in Bozeman for this project. Characterization was carried out at our MSU laboratories. After a brief description of the relevant material parameters, results for specific materials are presented.

Among the general material device parameters studied were:

- a) limits on the optical homogeneous line widths These limits arise from
  - population lifetime T<sub>1</sub>
  - crystal composition through nuclear and electron spin magnetic effects
  - phonon-induced relaxation
  - lattice structure through the effect of phonons
  - possible disorder modes
  - crystal field level gaps above the lowest energy levels these gaps affect phonon induced dephasing and
  - laser power-induced dephasing or 'instantaneous spectral diffusion',
- b) spin-lattice relaxation and spectral diffusion processes affecting spectral-hole lifetime,
- c) limits on hole-writing and reading rates,
- d) spectral hole lifetimes arising from metastable intermediate state lifetimes important for population storage mechanism, for example in Tm<sup>3+</sup> compounds,
- e) concentration-dependent dephasing and instantaneous spectral diffusion,
- f) growth-dependent effects arising from oxygen vacancies and paramagnetic impurities,
- g) material band gaps and position of ionic ground and excited states within the band gap for design of photon gating strategies (by ultraviolet and x-ray photoemission spectroscopy), and
- h) two-laser photon gated hole burning efficiencies and hole widths.

In addition to the materials prepared by Scientific Materials Corporation of Bozeman, we have studied samples from IBM (Roger Macfarlane), several groups at AT&T Bell Laboratories, Yale (W. P. Wolf, S. Mroczkowski), U. Lyon-France (B. Jacquier), Lawrence Livermore National Laboratory (M. J. Weber), Lawrence Berkeley Laboratory (N. M. Edelstein), M.I.T., CREOL, and others. Several unusual materials doped with rare earth and transition metal ions were obtained. Studies of Eu<sup>3+</sup>:Y<sub>2</sub>O<sub>3</sub> in carbon nanotubes were begun.

## Studies of Fundamental Limits on Performance:

As a first step in design and production of the optimum material for specific applications, it is important to understand the mechanisms that establish the fundamental limits on performance. Then tradeoffs in material design may be considered.

In applications of time-domain optical signal processing and memory devices, long optical coherence times (dephasing times)  $T_2$  can be especially important because this time largely determines the length of the data train that can be processed or stored at a fixed frequency and also affects the memory storage capacity in a swept carrier configuration.

#### Silicate Materials for Long Coherence Times

Our accomplishments include publication of studies on Eu<sup>3+</sup>, Pr<sup>3+</sup>, and Er<sup>3+</sup>-doped yttrium silicate materials. These have significantly improved homogeneous optical coherence times – record values for each ion. These silicate materials thus establish the standards by which all other materials are measured. We made significant improvements on several known systems containing Eu<sup>3+</sup> and Pr<sup>3+</sup> and made the first characterization of new systems containing Er<sup>3+</sup> and Tm<sup>3+</sup>.

Specific results are described for each material:

- For Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> we observed dephasing times of  $T_2 = 2.6$  ms, corresponding to a homogeneous linewidth of  $\Gamma_h = 122$  Hz optical linewidths and optical  $Q > 4 \times 10^{12}$ .

  This is the sharpest optical line ever observed in any solid of any type. The potential storage density enhancement factor is  $\Gamma_{inh}/\Gamma_h$ , = 10<sup>7</sup>, the ratio of inhomogeneous linewidth to homogeneous linewidth. These materials also have the potential for multi-GHz data rates. This material has been used by the Kachru group at SRI to demonstrate holographic optical storage at 10<sup>-8</sup> bit-error rates. It is also being used in the Templex Technologies optical dynamic random access memory (ODRAM). We are also developing it to provide stable lasers and optical frequency standards. To optimize this material for these applications, our studies on this material have been extended to higher Eu<sup>3+</sup> concentrations and to higher temperatures.
- b) We completed and published an extensive study of photon echo dynamics and energy levels for Pr<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> and have shown that this is the best available host for Pr<sup>3+</sup>.
- We studied Tm<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> and Tm<sup>3+</sup>:Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> in a search for comparable performance at diode laser wavelengths near 795 nm, but found that these hosts were not the best ones for Tm<sup>3+</sup>. In each case the homogeneous linewidth Γ<sub>h</sub> increased rapidly with temperature. The Tm<sup>3+</sup>:Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> was prepared especially for our studies by the Clarendon Laboratory,

University of Oxford, UK; those crystals did provide signal bandwidths as high as 100 GHz but it was very sensitive to temperature in the helium range. The reasons for the disappointing behavior of these silicates with Tm³+ were studied in detail, so that appropriate strategies could be devised to avoid these problems. In each case, spectroscopic studies revealed that small crystal field level gaps occurred just above the lowest level; these small gaps, together with the rapid increase of the density of lowenergy phonons with increasing temperature, provided the mechanism for phonon-induced dephasing.

More recently, we characterized  $Er^{3+}$ :  $Y_2SiO_5$  materials and obtained an optical dephasing time  $T_2 = 1.9$  ms corresponding to 170 Hz optical linewidth. This is a spectacularly narrow linewidth for a paramagnetic ion like  $Er^{3+}$ . Further discussion of  $Er^{3+}$  materials will

be given in a later section.

e) We contributed to understanding of fundamental limits on memory readout mechanisms. For example, we determined the mechanisms limiting homogeneous dephasing times in the best materials now available, we observed an important electromagnetic "shielding" phenomenon that minimizes loss of coherence, and we studied interactions between the "reading" light beam and the optical material (the so-called "instantaneous spectral diffusion effect" or laser-induced spectral diffusion).

## Other Oxides with Long Coherence Times - Eu3+:Y2O3, Tm3+:Y2O3, and Er3+:Y2O3

We used our refined photon echo decay measurement capabilities to study the well-known  $Eu^{3+}:Y_2O_3$  material and found that its dephasing properties are more ideal than those found in earlier measurement by several investigators. That is, laser-induced spectral diffusion ("instantaneous spectral diffusion" limited the values obtained by earlier workers. Growth-dependent dephasing still prevents this material from displacing  $Eu^{3+}:Y_2SiO_5$  in current applications, but the excellent thermal conductivity of  $Y_2O_3$  warrants additional effort to refine crystal growth. These materials are prepared under difficult conditions at temperatures near  $2000^{\circ}$  C where platinum crucibles can melt, and the excess dephasing appears to arise from structural disorder effects apparently associated with oxygen vacancies.

The  $Tm^{3+}$ : $Y_2O_3$  system has the potential to give the ideal properties sought in the  $Tm^{3+}$ -doped silicates, but  $Y_2O_3$  preparation is challenging. Photon echo studies of  $Tm^{3+}$ : $Y_2O_3$  were made using specially grown crystals. Two pulse photon echo decay measurements exhibited slightly non-exponential behavior. Application of a 440 Gauss magnetic field gave some narrowing to  $\Gamma_h = 11$  kHz. The crystal field levels gaps should not give significant temperature dependence in the helium range; nevertheless, a temperature dependence was measured, suggesting that structural disorder is present in the host crystal lattice. This is most likely due to two-level systems arising from oxygen vacancies, as observed for  $Eu^{3+}$ : $Y_2O_3$  by Flynn et al. Further development of  $Tm^{3+}$ : $Y_2O_3$  crystal growth is warranted to eliminate this problem.

The 1.5 micron transitions of  $Er^{3+}$ :  $Y_2O_3$  provide a very interesting group of properties as discussed further in the section on  $Er^{3+}$  materials. Realistic magnetic field operating conditions for  $Er^{3+}$  are provided by small button-sized permanent magnets. Homogeneous widths to  $\Gamma_b = 150$  Hz have been measured in our experiments.

#### Tm<sup>3+</sup> Materials for Coherent Transient Signal Processing Near 790 – 800 nm

Material design has been optimized for real time signal processing at semiconductor diode laser wavelengths in the 790-800 nm range. Particular effort was concentrated on eight  $Tm^{3+}$  compounds optimized for stimulated photon echo signal processing. These  $Tm^{3+}$  materials use the  ${}^3H_6 \leftrightarrow {}^3H_4$  transition of the  $Tm^{3+}$  ion. The temperature dependence of the dynamic properties depend strongly on the crystal lattice structure. High 3%  $Tm^{3+}$  concentration was investigated. A Coherent 899-21 Ti:Sapphire laser was used in our experiments to mimic diode lasers operating at a variety of wavelengths. Photon echo decays were measured as a function of temperature and magnetic field. Studies as a function of laser intensity ( $Tm^{3+}$  excitation density) explored the instantaneous spectral diffusion.

Macfarlane characterized the first Tm<sup>3+</sup> materials, focusing on Tm<sup>3+</sup>:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> (Tm<sup>3+</sup>:YAG) and contrasting its behavior to that of Tm<sup>3+</sup>:LaF<sub>3</sub>. The large fluorine nuclear magnetic moment resulted in shorter optical coherence times for LaF<sub>3</sub>, as expected. For Tm<sup>3+</sup>, as for Eu<sup>3+</sup> and Pr<sup>3+</sup>, it was thus natural to focus attention on oxides rather than fluorides.

This 795 nm diode laser spectral region has been used for demonstration of record areal storage density by the Mossberg Group using the swept carrier SHB technique; that demonstration used Tm<sup>3+</sup>:YAG. The Tm<sup>3+</sup>:YAG system has also been used for many signal processing devices by the Mossberg Group and Babbitt Group.

The first new Tm<sup>3+</sup> materials that we designed and characterized were the two silicates discussed above. Whereas the silicates are the best materials for Eu<sup>3+</sup> and Pr<sup>3+</sup> materials, they turned out to be less than ideal for Tm<sup>3+</sup>. "Accidental" near-degeneracies of crystal field levels in the ground and excited electronic states in both Y<sub>2</sub>SiO<sub>5</sub> and Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> led to unfavorable temperature dependence of the coherence properties. Low energy phonons, populated at low temperature, bridge the gaps between these low-lying electronic states, leading to phonon-induced optical dephasing.

That work led to the design and characterization of a series of garnet compounds, all of which have the same type of crystal structure but with different constituent ions, different lattice constants, and different internal strains. The compounds were: Tm³+:Y₃Ga₅O₁₂ (yttrium gallium garnet or YGG), Tm³+:Lu₃Al₅O₁₂ (lutecium aluminum garnet or LuAG), Tm³+:Y₁.₅Lu₁.₅Al₅O₁₂ (mixed yttrium-lutecium aluminum garnet or YLuAg), Tm³+:Y₂O₃ (yttria), and Tm³+:YAlO₃. We also made more extensive studies of Tm³+:Y₃Al₅O₁₂ (Tm³+:YAG), elucidating several new aspects of its behavior.

a) For 0.1% and 1%  $Tm^{3+}$ :YGG, a significant increase in the inhomogeneous linewidth to  $\Gamma_{inh} = 60$  GHz was achieved while maintaining a very favorable ratio of inhomogeneous to homogeneous linewidths of  $\Gamma_{inh}/\Gamma_h = 8 \times 10^7$ . At the same time, the effects of instantaneous spectral diffusion (laser-induced dephasing) were reduced for "one beam" experiments. The low measured oscillator strength on the transition of interest, however, meant that  $Tm^{3+}$ :YGG required nearly ten times higher laser power in device applications.

b) For 2%  $Tm^{3+}$ :LuAG, the first demonstration was made of operation at high  $Tm^{3+}$  concentration. The inhomogeneous linewidth was  $\Gamma_{inh} = 20$  GHz, and there was a very favorable ratio of inhomogeneous to homogeneous linewidths of  $\Gamma_{inh}/\Gamma_h = 8 \times 10^6$  despite the high Tm concentration.

c) To significantly increase the potential signal bandwidth compared to Tm³+:YAG, we designed the related mixed garnet compound Tm³+:YLuAG. Inhomogeneous broadening was increased by intentionally introducing structural disorder in the occupation of the yttrium sites with a 50:50 mix of Y³+ and Lu³+. This produced the substantially increased inhomogeneous broadening as expected, and there was no penalty on the ratio of inhomogeneous to homogeneous linewidths of Γ<sub>inh</sub>/Γ<sub>h</sub> = 6 x 10<sup>7</sup>. The oscillator strength of Tm³+:YLuAG was comparable to that of YAG. This favorable set of parameters extends the potential signal processing bandwidth at semiconductor laser wavelengths to 270 GHz. Remarkably, this material exhibited excellent coherence properties at 3% Tm concentration, and it has a strong optical absorption coefficient that will allow thin media (< 2 mm) for storage or processing devices.</p>

d) The  $Tm^{3+}$ :  $Y_2O_3$  system, as described above, did not produce the expected long dephasing times and narrow homogeneous linewidths  $\Gamma_h$ , even though the crystal field level spacings were relatively large. There was evidence of structural disorder on the oxygen sites. Further development of  $Tm^{3+}$ :  $Y_2O_3$  crystal growth is warranted.

e) Surprisingly, the Tm<sup>3+</sup>:YAlO<sub>3</sub> system did not produce observable photon echoes, using an apparatus that gave strong signals on other Tm<sup>3+</sup> systems on the same day in the same cryostat. This behavior is attributed to another case of "accidental" unfavorably close spacing of the crystal field energy levels – as for the silicates.

f) In Tm:YAG, we investigated the effects of Tm concentration with specific device applications in mind. With optimized concentration, this material can play a major role in demonstration of a variety of signal processing capabilities.

In conclusion, we studied  $Tm^{3+}$  in  $Y_2SiO_5$  (YSO),  $Y_2Si_2O_7$ ,  $Y_2O_3$ ,  $Y_4O_3$ ,  $Y_3AI_5O_{12}$  (YAG),  $Y_3Ga_5O_{12}$  (YGG),  $Lu_3AI_5O_{12}$  (LuAG), and the mixed garnet system  $Y_{1.5}Lu_{1.5}AI_5O_{12}$  (YLuAG) using photon echoes. The temperature-dependent dynamic properties depend strongly on lattice structure. Measured dephasing times were in the 100  $\mu$ s range, except for YSO and YAIO<sub>3</sub>, where dephasing was much faster due to direct phonon processes involving the especially small energy gaps between low lying electronic states of the  $^3H_6$  crystal field manifold in those compounds. In most of these studies, particularly the earlier ones, Tm ions were doped into these crystals in low concentration (~0.1%) to minimize the  $Tm^{3+}$ - $Tm^{3+}$  interaction. Once it was evident that laser-induced instantaneous spectral diffusion was a small effect in these materials, higher concentrations were investigated for  $Tm^{3+}$ :YGG and  $Tm^{3+}$ :YAG. Similar insensitivity to concentration in 3%  $Tm^{3+}$ :YLuAG allowed narrow 4.6 kHz homogeneous linewidths to be obtained simultaneously with a very broad inhomogeneous width of 270 GHz. This material is especially interesting for signal processing applications. It provides a ratio  $\Gamma_{inh}/\Gamma_h \sim 10^8$ .

In the garnets the two pulse photon echo decays were mostly non-exponential due to frozen core effects of the Al spins.

Basic spectroscopic results for the Tm<sup>3+</sup> materials are summarized in the table below.

Crystal	Tm concentration	Crystal Symmetry	Site Symmetry	Wavelength (nm)
Y <sub>3</sub> Al <sub>5</sub> O <sub>12</sub>	0.1%, 1%, 2%, 3.3%, 4%	O <sub>h</sub>	$D_2$	793.156
Lu <sub>3</sub> Al <sub>5</sub> O <sub>12</sub>	2%	O <sub>h</sub>	$D_2$	793.808
Y <sub>1.5</sub> Lu <sub>1.5</sub> Al <sub>5</sub> O <sub>12</sub>	3%	Oh	$D_2$	793.527
Y <sub>3</sub> Ga <sub>5</sub> O <sub>12</sub>	0.1%, 1%	O <sub>h</sub>	$D_2$	795.108
Y <sub>2</sub> O <sub>3</sub>	0.1%	T <sub>h</sub> 7	$C_2$ , $C_{3i}$	796.286
Y <sub>2</sub> SiO <sub>5</sub>	0.1%	C <sub>2h</sub> <sup>6</sup>	C <sub>1</sub>	790.781 790.769
Y <sub>2</sub> Si <sub>2</sub> O <sub>7</sub>	0.1%		C <sub>1</sub>	790.427
YAlO <sub>3</sub>	0.1%, 4%	D <sub>2h</sub> <sup>16</sup>	Cs	798.845

Dynamic properties for the Tm<sup>3+</sup> materials are summarized in the table below, both for zero applied magnetic field and in weak fields. Exponential decays were observed for Tm<sup>3+</sup>:Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>. Nonexponential decays were fitted to the standard Mims expresssion for the other materials.

Crystal	Γ <sub>inh</sub> (GHz)	T <sub>M</sub> (μs)	x	$\Gamma_{ m inh}/\Gamma_{ m h}$	T <sub>M</sub> (μs)	x	$\Gamma_{\rm inh}/\Gamma_{\rm h}~(10^6)$
Magnetic Field		B = 0			B > 100 G		
$Y_3Al_5O_{12}$	20	116	2.1	$7 \times 10^6$	130	1.3	8 x 10 <sup>6</sup>
Lu <sub>3</sub> Al <sub>5</sub> O <sub>12</sub>	20	90	1.3	6 x 10 <sup>6</sup>	120	1.0	$7 \times 10^6$
Y <sub>1.5</sub> Lu <sub>1.5</sub> Al <sub>5</sub> O <sub>12</sub>	270	55	1.1	$4.6 \times 10^7$	75	1.0	6 x 10 <sup>7</sup>
Y <sub>3</sub> Ga <sub>5</sub> O <sub>12</sub>	55	326	1.65	$5.5 \times 10^7$	460	1.2	8 x 10 <sup>7</sup>
Y <sub>2</sub> O <sub>3</sub>	3.6	19	1.15	2 x 10 <sup>5</sup>	21.4	1.0	$2.5 \times 10^5$
Y <sub>2</sub> SiO <sub>5</sub>	2.6 4.5	4.8 1.3	1.0 1.0	$4 \times 10^4$ $2 \times 10^4$	No change		
Y <sub>2</sub> Si <sub>2</sub> O <sub>7</sub>	100	T <sub>2</sub> =	23 μs	7 x 10 <sup>6</sup>	No change		hange
YAlO <sub>3</sub>	Unfavorable Energy Level Splittings						

With these Tm<sup>3+</sup> materials we have again established new ranges of parameter space for device applications with semiconductor lasers -- they could handle signal bandwidths as high as 270 GHz.

These Tm<sup>3+</sup> materials also have given us new insights into laser-induced instantaneous spectral diffusion (laser-induced dephasing).

# $Er^{3+}$ Materials for All-Optical Data Switching and Routing, Buffer Memories, and other Network, Memory, Optical Interconnect, and Signal Processing Devices in the 1.5 $\mu m$ Communication Band

Particular effort during the third year was concentrated on Er<sup>3+</sup> compounds for SHB applications with semiconductor diode lasers in the particularly important 1.5 µm communication band. The broad goal of this development work is to provide materials that enable all-optical data switching and routing, buffer memories, and other network, memory, optical interconnect, and signal processing devices operating at multi-terabit/s rates and to provide highly parallel data transmission. The specific material requirement was that the materials be developed to allow these Er<sup>3+</sup> systems to interface with existing technology in the 1.5 µm communications band.

Our photon echo characterization study of Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> was the first report of a practical material for applications in this spectral region, and it was published in Optics Letters and presented at CLEO'97. As a further part of this project, we have already demonstrated a key element of an optical packet routing system exploited this material (submitted to Optics Letters). The Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> material is also being used for demonstrations of other signal-processor, memory, and router applications.

Desirable features for data routing and processing and for buffer memories include long  $T_2$  for storing or processing long pulse trains, low spectral diffusion, and convenient wavelengths. To obtain long  $T_2$  (corresponding to narrow  $\Gamma_h$ ), a magnetic field is needed to split the Kramers doublet  $Er^{3^+}$  electronic levels and minimize spin-flip-induced dephasing for photon echo and hole burning applications. It has been possible to achieve realistic values of  $T_2$  under simple constraints as noted above using the permanent magnets.

Three illustrations demonstrate that excellent performance can be obtained under practical device conditions:

- 1. Small button-sized Nd-Fe-B permanent magnets provided 2.5 kG at the 0.001%  $Er^{3+}$ : Y<sub>2</sub>SiO<sub>5</sub> crystal and gave  $\Gamma_h \sim 2.5$  kHz. These experimental conditions are well suited to the device demonstrations that we have performed and which we describe below.
- 2. For 0.005%  $\rm Er^{3+}: Y_2O_3$  at zero magnetic field, strong echoes, 3 % as intense as the exciting laser pulse, and  $\rm T_2 \sim 9~\mu s$  and  $\rm \Gamma_{inh} = 35~kHz$  were observed.
- 3. In 0.06%  $Er^{3+}$ :LiNbO<sub>3</sub>, inhomogeneous linewidth  $\Gamma_{inh}$  = 250 GHz and  $\Gamma_h$  = 4 kHz at 5 kG. That makes this an interesting material for spatial-spectral holographic applications where the ratio of the inhomogeneous to homogeneous linewidth  $\Gamma_{inh}/\Gamma_h$  is important.

Optical dephasing and spectral diffusion have been studied for the  ${}^4I_{15/2} \leftrightarrow {}^4I_{13/2}$  transition of Er<sup>3+</sup> at 1.5 µm in seven crystal systems: Y<sub>2</sub>SiO<sub>5</sub>, Y<sub>2</sub>O<sub>3</sub>, LiNbO<sub>3</sub>, YAG, YAlO<sub>3</sub>, CaWO<sub>4</sub>, and SrWO<sub>4</sub>. Our motivation for choosing these materials is described below. Photon echo decay measurements were used to determine optical dephasing times T<sub>2</sub> as a function of applied magnetic field and temperature; stimulated photon echo decays were used to study spectral diffusion.

A tunable diode laser was constructed in our laboratory for these photon measurements, using an external cavity design. That laser produces 7 mW of single frequency output, a value larger than commercial lasers provided at the time. Our laser is capable of operation at all communication wavelengths. An erbium-doped fiber amplifier was used to amplify this diode laser's output to  $\sim$ 50 mW for the two-pulse and three-pulse photon echo experiments. This system also enabled us to make high resolution determinations of the inhomogeneous widths  $\Gamma_{\rm inh}$  for each material.

The  $\mathrm{Er}^{3^+}$  ions were doped into each crystal at very low concentration (~ 10 parts per million) to minimize dephasing and spectral diffusion caused by  $\mathrm{Er}^{3^+}$ - $\mathrm{Er}^{3^+}$  interactions; those range-dependent interactions lead to mutual electronic spin flips of the  $\mathrm{Er}^{3^+}$  ions. Our immediate goal was elucidation of the important differences in performance of the various host crystals. Additional studies will explore the practical upper limits of  $\mathrm{Er}^{3^+}$  concentration.

The narrowest homogeneous linewidth that we have observed is  $\Gamma_h \sim 150$  Hz, which establishes a dramatic new record for Kramers ions and closely rivals our record value for  $\Gamma_h$  in Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>. This 150 Hz value was measured in 0.001% Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> at 80 kG and 1.5K. Based on the long lifetime of the  $^4I_{13/2}$  level in these crystals, the ultimate value of  $\Gamma_h$  might reach 12 Hz.

So far, the holeburning mechanism in these seven  $Er^{3+}$  systems is two-level saturation, with hole lifetimes determined by the upper electronic state lifetime ( $T_1 \sim 13$  ms). We are now investigating several new materials. including deuterated  $Er^{3+}$ :CaF<sub>2</sub>, that offer persistent hole burning and the prospect of photon gating.

All of the crystals that we have studied in detail were grown by Scientific Materials except for Er<sup>3+</sup>:LiNbO<sub>3</sub>. Crystal design was a joint effort of our laboratory and Scientific Materials.

- Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> was chosen, as noted earlier, based on our previous demonstration of ideal performance in this host for Eu<sup>3+</sup> and Pr<sup>3+</sup> ions. With its low nuclear magnetism, it was expected to exhibit very long dephasing times, and this goal was realized.
- Er<sup>3+</sup>:Y<sub>2</sub>O<sub>3</sub> was chosen to eliminate nuclear magnetism and also because it has very ideal thermal properties.
- Er<sup>3+</sup>:YAG was studied, based on the success of YAG as a host for Tm<sup>3+</sup>, as described earlier, and based on its wide practical use as a high power laser material.
- Er<sup>3+</sup>:YAlO<sub>3</sub> was expected to be similar to YAG in many ways, but it has polarized transitions and other different optical properties.
- Er<sup>3+</sup>:CaWO<sub>4</sub> and Er<sup>3+</sup>:SrWO<sub>4</sub> were also chosen to eliminate nuclear magnetism. The need for charge compensation distinguishes these materials from the others listed above.
- Er<sup>3+</sup>:LiNbO<sub>3</sub> was chosen based on its wide use as a nonlinear optical material.

The table below summarizes basic spectroscopic properties of these materials:

Crystal	Er <sup>3+</sup> Concentration	Crystal Symmetry	Site Symmetry	Wavelength
Y <sub>2</sub> SiO <sub>5</sub>	0.001%, 0.005%	C 2 h	C <sub>1</sub>	1536.14 nm site 1
			C <sub>1</sub>	1538.57 nm site 2
Y <sub>2</sub> O <sub>3</sub>	0.005%	$T_h^7$	C <sub>2</sub>	1535.28 nm
LiNbO <sub>3</sub>	0.06%	$C_{3v}^6$	C <sub>3</sub>	1531.52 nm
YAlO <sub>3</sub>	0.005%	$D_{2h}^{16}$	Cs	1514.38 nm
CaWO <sub>4</sub>	0.005%	$C_{4h}^6$	S <sub>4</sub>	1532.30 nm
SrWO <sub>4</sub>	0.005%, 0.05%, and 0.5%	C4h	S <sub>4</sub>	1533.55 nm
YAG	0.1%	O <sub>h</sub>	D <sub>2</sub>	1526.97 nm

Our studies of Er<sup>3+</sup>: Y<sub>2</sub>SiO<sub>5</sub> have elucidated the important dephasing mechanisms for Er<sup>3+</sup>. The conclusions of this pioneering analysis of the Er<sup>3+</sup> ion will also apply to other Kramers-ion systems (ions with an odd number of electrons) like Nd<sup>3+</sup>, Sm<sup>3+</sup>, Dy<sup>3+</sup>, or Yb<sup>3+</sup>.

The dominant dephasing mechanism in all of the Er<sup>3+</sup> materials listed above was found to be Er<sup>3+</sup>-Er<sup>3+</sup> mutual electronic spin flips. The spin flip rate depends on Er<sup>3+</sup> concentration and on the populations of the spin sublevels – those populations in turn depend on temperature and external magnetic fields. The contribution of this mechanism in our extremely dilute samples spans the extreme range of from 100 Hz to 100 kHz, and it would be far larger at increased Er<sup>3+</sup> concentration. A weaker dephasing mechanism involves Y nuclear spin flips, which we have also shown to be important for Eu3+ and Pr3+ ions. In the materials that have nuclei with larger nuclear magnetic moments, such as Al<sup>3+</sup> in YAG, the nuclear spin flip dephasing contribution is more important than in YSO, but it is still second in importance to the Er-Er spin flips. The Y<sup>3+</sup> nuclear spins are largely frozen by the magnetic fields arising from the Er3+ electronic spins, but this mechanism contributes at long times an amount < 1 kHz in Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>. Electronic spin flips can be quenched by strong magnetic fields, but the nuclear magnetic dephasing mechanism cannot be quenched - the magnetic energies of the nuclear spins are much smaller than kT at any reasonable temperature even in the helium range. Phonon scattering and phonon absorption contribute less than 100 Hz at 1.6 K. Finally, the limiting contribution is the Er<sup>3+</sup> excited state population decay (T<sub>1</sub> process) which contributes only 12 Hz. By applying a strong 80 kG magnetic field in our experiments on Er3+: Y2SiO5, we have suppressed most of these contributions and have thus obtained the second narrowest optical linewidth observed in any solid – 150 Hz.

The difference between the predicted 12 Hz limit and our 150 Hz measurement suggests that subtle frequency jitter in the diode laser may be contributing some of the residual observed line width. These Er<sup>3+</sup> materials at 1.5 microns, like Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> in the visible, can thus serve a valuable role in laser diagnostics. Photon echo measurements on Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> currently provide the most critical test available for high quality diode lasers in the 1.5 micron region.

Specific performance data obtained from each of the systems studied is listed below. The  $T_M$  and x parameters are obtained from fits to the standard Mims expression for nonexponential decays. Both sites in YSO have similar dynamical properties.

		H <sub>0</sub> =0		High Field Values		
Material	Γ <sub>inh</sub> (GHz)	T <sub>M</sub> (μs)	x	T <sub>M</sub> (μs)	x	$\Gamma_{ m inh}$ / $\Gamma_{ m h}$
YSO 0.001%	0.5	3.3	1	2100	1.0	$3\cdot 10^6$
Y <sub>2</sub> O <sub>3</sub> 0.005%	1	18	1.85	105	1.4	3 · 10 <sup>5</sup>
LiNbO <sub>3</sub> 0.06%	200			170	2.0	1 · 10 <sup>8</sup>
YAlO <sub>3</sub> 0.005%	1			265	2.2	8 · 10 <sup>5</sup>
CaWO <sub>4</sub> 0.005%	1			140	1.5	4 · 10 <sup>5</sup>
SrWO <sub>4</sub> 0.05%	1			72	1.0	2 · 10 <sup>5</sup>
YAG 0.1%	30			75	1.5	7 · 10 <sup>6</sup>

In summary, in the course of these Er³+ studies, we have demonstrated the longest T₂ ever observed in a Kramers-ion system (odd number of electrons), the second narrowest line width ever observed in a solid. The primary dephasing mechanism was attributed to the Er³+-Er³+ mutual spin flips, but that can be controlled by inexpensive and compact permanent magnets. In the YAG system, the Al nuclear moment also plays a role in the optical dephasing. Spectral diffusion was observed in all the crystals. The diffusion rate is comparable to the estimated Er³+-Er³+ mutual spin flip rate. The Er³+:YSO provides the longest T₂, but to obtain other wavelengths or operating conditions, all but one of the studied materials are useful. The Er³+:SrWO₄ has a weak oscillator strength compared to Er³+:CaWO₄. In addition, the two materials Er³+:Y₂O₃ and Er³+:LiNbO₃ have particularly interesting features. Very strong echoes were observed for each. Echoes were observable in Er³+:Y₂O₃ without any applied magnetic field. The inhomogeneous line width of Er³+:LiNbO₃ offers 200 GHz bandwidth for signal processing applications.

With the wide range of materials described above, we have again established new ranges of parameter space for device applications, particularly with semiconductor lasers.

The eighth Er<sup>3+</sup> material that we studied was chosen to provide versatile optical frequency standards. For that purpose, we are investigating the persistent spectral hole burning materials Er<sup>3+</sup>:D<sup>-</sup>:CaF<sub>2</sub> and Tm<sup>3+</sup>:D<sup>-</sup>:CaF<sub>2</sub>. Frequency-locking of diode lasers to those materials is being investigated under another AFOSR grant.

# Device Demonstration: Address-Header Decoder Operating at 1536 nm using Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>

The frequency- and time-domain phenomena of spectral hole burning and photon echoes may be exploited together with the ideas of spatial holography in spatial-spectral holography (SSH) to obtain optical signal processing and storage capabilities that may be used separately or in powerful combinations. We have examined applications as well as materials in this project.

We recently submitted to Optics Letters our use of Er<sup>3+</sup>:YSO in a demonstration of address header decoding, one of several important components of high speed data routing by optical packet switching. The advantages of optical packet switching include fast data routing in the wavelength and space domains and transparency to packet bit rate and format. Specifically, we programmed a crystal to recognize bi-phase coded address headers and to decode an arbitrary sequence of these headers, resulting in spatially-discriminated optical output pulses. Each output pulse could be used to route or switch the optical data packet or the combined header and data packet in real time, requiring only a compact optical delay.

Materials and applications for a variety of other applications have been considered.

#### **New Gated Spectral Hole Burning Materials**

We have also explored new materials and materials concepts for photon gating of persistent spectral hole burning. The "robustness" and long-term persistence associated with photon gating are important to storage and processing and to frequency standard and optical clock applications.

In conjunction with Scientific Materials, we designed and have begun evaluation of doubly-doped materials with paired sites capable of stabilizing ions in multiple ionization states. Double doping of active ions that are chosen so that each can be stabilized in more than one ionization state might overcome the low efficiency problems for gating. This concept may also reduce the effect of gating on the surrounding area in the crystal and hence reduce broadening of previously written holes and consequent destruction of previously written data by spectral diffusion.

To extend knowledge of photon gating systems, we have studied materials where photoionization of Tb3+ to Tb4+ can open new areas of study and offer material systems for operation in the blue spectrum, where future development of optical storage systems is exected. Specifically, we have carried out spectroscopic studies of Tb3+:LiYF4, Tb3+:LuPO4, Tb3+:ScPO4, Tb3+:YPO4, Tb3+:YAG, Tb3+:Y2SiO5, Tb3+:Y2Si2O7, and Tb3+:YAlO3. Both visible and ultraviolet absorptions studies have been completed, along with some characterization of the excited state absorption spectra. There are a number of reasons for exploring terbium. The Tb<sup>3+</sup> ion has the lowest energy 4f to 5d transitions of any rare earth ion, so both the hole burning and gating could be carried out with visible light. The terbium ion also exists in both the Tb3+ and the Tb4+ ionization states. Prospects for photon gated hole burning in the terbium system have not been explored in any of the earlier studies of photon gated hole burning in rare earth materials. We had previously reported hyperfine hole burning in Tb3+:LiYF4 with hole lifetimes of minutes. Hole burning was observed during this project in Tb3+: YAlO3, and our present analysis indicates that these holes were persistent, indicating gating. In Tb3+:YAlO3, ultraviolet absorption spectra that we have recorded show that the second photon gating energy values are greater than first hole burning photon's <sup>7</sup>F<sub>6</sub> to <sup>5</sup>D<sub>4</sub> transition energy.

Experimental apparatus under development in our laboratory for gating studies includes photoconductivity and photoemission experiments.

#### Personnel Supported and Associated

Two postdoctoral fellows have worked on this AFOSR research project.

- Dr. Yongchen Sun joined the group from Professor Richard Meltzer's group at the University of Georgia in July 1, 1996, for a two-year postdoctoral appointment as an Assistant Research Professor.
- 2) Dr. Flurin Koenz won a postdoctoral fellowship from the Swiss National Science Foundation specifically for research in Cone's laboratory. Dr. Koenz arrived from Bern, Switzerland, on October 14, 1996, with all expenses paid by the Swiss NSF.

Cone's group has had the following graduate students involved in AFOSR research. Their salaries were paid by fellowships or by other research grants.

- Randy W. Equall, Ph. D. Completed May, 1995, on *Photon Echo Studies of Rare Earth Activated Materials for Optical Memory and Signal Processing Devices*. Received job offers from Intel Corporate Research Laboratory and Scientific Materials Corporation -- Currently at Scientific Materials Corporation, an AFOSR SBIR Contractor.
- Guangming Wang, Ph. D. Completed May, 1997, on Defect and Coherent Transient Optical Spectroscopy of Rare Earth Doped Crystals. He worked on symmetry aspects of hole burning materials and coherent transients. He is currently engaged in development of ultraviolet laser lithography for integrated circuits at Lithography Systems, Inc in Wilton CT.
- 3) Greg White, graduate student in physics.
- 4) Lane Seeley, graduate student in physics.
- Todd Harris, graduate student in physics, developed laser instrumentation at 1.5 μm. Todd constructed an external cavity diode laser (ECDL) and connected it to an erbium doped fiber amplfier (EDFA), and he participated in photon echo characterization of Er<sup>3+</sup> compounds for 1.5 μm devices.
- 6) Charles Thiel, graduate student in physics, worked on gated spectral hole burning in Tb<sup>3+</sup> materials and UV spectroscopy of other candidates.
- Sebastien Ermeneux, a graduate student in physics, arrived February 13, 1997, and began working on LiNbO<sub>3</sub> and other Er<sup>3+</sup> materials relevant to this project. We are also developing a 1.5 μm solid state laser.
- 8) Gregory Reinemer, a physics graduate student, was introduced to this work and to experimental techniques.
- 9) Another graduate student, Thomas Bottger, with experience in rare-earth doped semiconductors, has just joined the group.

Cone's group had nine undergraduate students involved in AFOSR research during this grant period.

- 1) Charles Thiel, undergraduate student in physics., B. S. in Physics, May, 1996, and a Congressional Goldwater Scholar. Charles has continued his studies for a Ph.D. in this group at Montana State University.
- 2) Paulo Rigg, undergraduate student in physics, B. S. in Physics, May, 1995.
- 3) Sean Dirkes, undergraduate in electrical engineering.
- 4) John Hill, undergraduate in electrical engineering technology.
- 5) Guy Santiglia, undergraduate in electrical engineering.
- Tyler Morgus, undergraduate student in physics, BS in Physics, May, 1996. Tyler entered the Ph.D. program at Lehigh University in September, 1996.
- 7) Kevin Yager, B.S. in Physics at MSU, 1997.
- 8) Glenn Omdahl, undergraduate student in physics, will receive BS in Physics, May, 1998, and begin work for AFOSR SBIR Contractor, Scientific Materials Corporation.
- 9) Seth Mayer, undergraduate student in physics.

- Dr. R. M. Macfarlane of IBM Almaden Research Center visited our group on
- 1) August 3-10, 1995, for experiments and to attend the Bozeman Workshop on Applications of Persistent Spectral Hole Burning
- 2) March 3 March 11, 1996, for discussions, experiments, and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning
- 3) October 26 30, 1996,
- 4) February 15 23, 1997,
- 5) August 8 14, 1997, for experiments, discussions, and to attend the AFOSR Workshop on Frontiers of Applications of Persistent Spectral Hole Burning.

Crystals of Tm<sup>3+</sup>:Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub> were grown specifically for our experiments by K. W. Godfrey and F. R. Wondre of the Crystal Growth Unit of the Clarendon Laboratory, University of Oxford.

Dr. M. J. M. Leask of the Clarendon Laboratory, University of Oxford, UK, visited Cone's group

- 1) September 4-21, 1994 for experiments and writing
- 2) October 4 December 4, 1995, while on sabbatical leave
- 3) February 29 March 7, 1996, for discussions, writing, and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning
- 4) August 6 20, 1996, for experiments on rare-earth nanocrystals in carbon nanotubes prepared at the University of Oxford.
- 5) February 14 26, 1997, for discussions, manuscript writing, and to attend the AFOSR Workshop on Frontiers of Applications of Persistent Spectral Hole Burning.

We are obtaining Er<sup>3+</sup> doped carbon nanostructures for characterization, in collaboration with Leask and Malcolm L. H. Green, FRS, at the Inorganic Chemistry Laboratory of the University of Oxford.

Dr. Peter Hansen visited from the University of Oxford, UK, from April 5-13, 1995.

There was regular electronic communication with both the IBM and Oxford groups throughout the year.

A third postdoctoral fellow, Nicholas Strickland from the group of Dr. Glynn Jones at University of Canterbury, Christchurch, New Zealand, joined Cone's group in Autumn, 1996, supported by a separate AFOSR project. The Glynn Jones' group is well known for its work on persistent spectral hole burning in H- and D-doped CaF<sub>2</sub> rare earth materials and for general expertise in rare earth spectroscopy and modeling.

Dr. W. R. Babbitt of the University of Washington visited in May, 1995, and presented a Physics Colloquium. Babbitt visited again on March 6-8, 1996, to discuss a joint University of Washington - Montana State University proposal on a prototype terabit/second optical fiber communications link using spectral hole burning concepts and frequency-division multiplexing. This proposal was submitted to the ARPA-Ultra-Photonics program in June, 1996.

Dr. Yongchen Sun of the University of Georgia Department of Physics visited February 29 - March 7, 1996, for discussions and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning, in preparation to assume a postdoctoral appointment here as "Assistant Research Professor."

Dr. Richard Meltzer of the University of Georgia visited February 29 - March 7, 1996, for discussions and to attend the Big Sky Workshop on Applications of Persistent Spectral Hole Burning.

Hai Lin of the University of Oregon, Jean-Pierre Galaup of Paris, Aleksander Rebane of ETH-Zurich, and Yiping Zhang of SRI, Menlo Park, toured Cone's research laboratory in conjunction with the Big Sky Workshop on Applications of Persistent Spectral Hole Burning, March, 1996.

Dr. Bernard Jacquier from Universite Lyon I visited July 27 - August 3, 1996, to discuss hole burning in rare earth doped optical waveguides and dielectric microcavities.

The interdisciplinary MSU Optical Technology Center (OpTeC) group submitted an NSF Optical Science and Engineering proposal on hole burning memory materials. This group consists of Cone and Carlsten from Physics, Singel and Spangler from Chemistry, and Cady from Electrical Engineering. Dr. Aleksander Rebane has accepted a position in the group.

Cone, Carlsten, and Rebane submitted an NSF Major Research Instrumentation (MRI) proposal on hole burning memory materials. Cone and W. R. Babbitt prepared an NSF research proposal on multi-terabit/s frequency-domain multiplexing at 1.5 µm. Cone, Carlsten, Babbitt, and Rebane are all members of the interdisciplinary MSU Optical Technology Center (OpTeC) group. The OpTeC group of ten faculty in Chemistry, and Electrical Engineering is preparing an NSF Materials Research Science and Engineering Center (MRSEC) proposal on hole burning memory materials.

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   W. Equall, R. L. Cone, and R. M. Macfarlane, Phys. Rev. B 52, 3963-3969 (1995).
- 2. Ultra-slow dephasing and dephasing mechanisms in rare earth materials for optical data storage, R. L. Cone, R. W. Equall, Y. Sun, R. M. Macfarlane, and Ralph Hutcheson, Laser Physics 5, 573 (1995).
- 3. The hyperfine splitting of the ground state and first excited state of holmium in yttrium hydroxide, D. St. P. Bunbury, C. Carboni, R. L. Cone, M. A. H. McCausland, S. Mroczkowski, and J. W. Ross, J. Phys. Condensed Matter 7, 6125-6136 (1995).
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- 5. Optical Dephasing Mechanisms in Tm<sup>3+</sup>:Y<sub>2</sub>Si<sub>2</sub>O<sub>7</sub>, G. M. Wang, R. W. Equall, R. L. Cone, M. J. M. Leask, K. W. Godfrey, and F. R. Wondre, Opt. Lett. 21, 818 (1996).
- 6. Spectral Holeburning of Eu<sup>2+</sup> in CaF<sub>2</sub>, D. M. Boye, R.M. Macfarlane, Y. Sun, and R. S. Meltzer, Phys. Rev. B. **54**, 6263 (1996).
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- 9. Spectral Hole Burning and ODNQR in Flux-Grown Stoichiometric Europium Vanadate Crystals, P. C. Hansen, M. J. M. Leask, B. M. Wanklyn, Y. Sun, R. L. Cone, and M. M. Abraham, Phys. Rev. B 46, 7918-7929 (1997).
- 10. Demonstration of Real-Time Address Header Decoding for Optical Data Routing at 1536 nm, T. L. Harris, Y. Sun, R. L. Cone, R. M. Macfarlane, and R. W. Equall, submitted to Optics Letters, November 5, 1997.
- 11. Angle-Dependent Optically Detected Nuclear Magnetic Resonance on the Defects in EuVO<sub>4</sub>, Y. Sun, R. L. Cone, M. J. M. Leask, and M. M. Abraham, manuscript prepared for Phys. Rev. B.

- 12. Spectroscopic Properties of Tm<sup>3+</sup>-Doped Materials for Coherent Transient Signal Processing near 795 nm, Y. Sun, G. M. Wang, R. L. Cone, R. W. Equall, and R. M. Macfarlane, manuscript prepared for Phys. Rev. B.
- 13. Hyperfine Interactions and Site Symmetry for Intrinsic and Defect Sites of Pr<sup>3+</sup> in Yttrium Aluminum Garnet, G. M. Wang, Y. Sun, R. W. Equall, R. L. Cone, R. M. Macfarlane, and M. J. M. Leask, manuscript prepared for Phys. Rev. B.
- 14. Photon Echo Studies of Optical Dephasing in Er<sup>3+</sup>-Doped Materials for All-Optical Switching and Routing at 1.5 microns, Y. Sun, T. L. Harris, R. L. Cone, R. M. Macfarlane, and R. W. Equall, manuscript prepared for Phys. Rev. B.

#### **Interactions**

#### Presentations and Participation at Meetings and Conferences

AFOSR Workshop Organizer: Material Requirements for Persistent Spectral Hole Burning and Time-Domain Optical Storage and Processing, Bozeman, MT, August 3-4, 1995, plus participation as follows:

- a) "Materials Overview," R. L. Cone
- b) "Material Characterization at MSU," R. W. Equall, G. M. Wang, R. L. Cone, and R. M. Macfarlane,
- c) "Panel Discussion on Spectral Diffusion," R. L. Cone.

Invited conference talk, Ultraslow Optical Dephasing and Materials for Photon Echo and Spectral Hole Burning Memories, R. L. Cone, 1994 Annual Meeting of the Optical Society of America, Dallas, Texas, October, 2-7, 1994.

Invited conference talk, Ultraslow Dephasing and Dephasing Mechanisms in Rare Earth Materials for Optical Data Storage, R. L. Cone, International Workshop on Laser Physics entitled Transient Coherent Phenomena honoring Professor Sven Hartmann, discoverer of the photon echo, Columbia University and City College of New York, New York, New York, October 10-14, 1994.

Invited talk, Ultra-Slow Dephasing and Dephasing Mechanisms in Rare Earth Materials for Optical Data Storage, R. W. Equall, Intel Research Laboratory, Portland, Oregon, November, 1994.

Rufus Cone attended Advanced Solid-State Lasers Conference, Memphis, Tennessee, January 30-31, 1995.

Invited talk, Materials for Time-Domain Optical Storage, R. L. Cone, AFOSR Workshop on Persistent Spectral Hole Burning, University of Washington, Seattle, Washington, February 1-2, 1995.

Contributed conference talk, Homogeneous Broadening of Optical Transitions in Pr<sup>3+</sup> and Tm<sup>3+</sup> doped Y<sub>2</sub>SiO<sub>5</sub>, R. W. Equall, R. L. Cone, R. M. Macfarlane, and R. L. Hutcheson, APS March Meeting, San Jose, California, March 20-24, 1995.

"New Materials for Persistent Spectral Hole Burning Optical Memories and Signal Processing Applications," R. W. Equall, MSU Physics Department Colloquium, October 20, 1995.

R. L. Cone attended the Advanced Solid-State Lasers meeting, San Francisco, CA, January 31-February 2, 1996.

R. L. Cone attended the American Physical Society 1996 March Meeting, St. Louis, MO, March 18-22, 1996.

Invited talk, Hole Burning Materials and Physical Processes, R. L. Cone, AFOSR Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, March 3-6, 1996.

Invited talk, Spectral-Hole-Burning Frequency Standards and Stable Lasers, R. L. Cone, AFOSR Contractors Workshop on Time and Frequency Standards, Phillips Laboratory, Kirtland Air Force Base, Albuquerque, NM, April 25, 1996.

Program Committee, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

Presider, Session on Inorganic Optical Materials, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

Refereed conference talk, Tm Materials for Coherent Transient Optical Memories and Signal Processors, R. W. Equall, G. M. Wang, G. A. White, R. L. Cone, M. J. M. Leask, R. M. Macfarlane, and R. L. Hutcheson, Conference on Lasers and Electro-Optics (CLEO/QUELS '96), Anaheim, CA, June 2-7, 1996.

Invited conference talk, Nonlinear Optical Studies of Rare Earth Compounds for Optical Storage and Signal Processing and for Lasers, R. L. Cone, 21st Rare Earth Research Conference, Duluth, MN, July 7-12, 1996.

Invited conference talk, Photon Echo Measurements of Optical Dephasing in Rare-Earth Doped Insulators, R. L. Cone, International Conference on Luminescence, Prague, Czech Republic, August 18-23, 1996.

Montana State University "OpTeC" Conference on Optical Science and Laser Technology, Bozeman, MT, Sept. 5-6, 1995.

Invited talk, Rare Earth Crystals for Solid State Lasers and for Optical Data Storage and Signal Processing, R. L. Cone.

Contributed talk, Tm<sup>3+</sup>-Doped Materials for Optical Memory and Signal Processing Applications, Guangming Wang, graduate student.

Contributed talk, Materials for Photon Gated Hole Burning, G. White, graduate student. Contributed talk, Searches for Tb Compounds Exhibiting Photon Gated Hole Burning, C. W. Thiel, undergraduate student.

Invited Conference Talk, Materials for Coherent Transient Optical Memories and Signal Processors, R. L. Cone, 5th International Meeting on Hole Burning and Related Spectroscopies, Brainerd, MN September 13-17, 1996.

Invited Conference Talk, Materials and Material Concepts for Devices at Infrared, Visible, and Communication Wavelengths Based on Ions in Crystals, R. L. Cone, Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, February 16-19, 1997.

Refereed Conference Talk, Measurement of Photon Echoes at 1.5 microns in Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> using a Diode Laser and Amplifier, R. M. Macfarlane, T. L. Harris, Y. Sun, R. L. Cone, and R. W. Equall, Quantum Electronics and Laser Science Conference (CLEO/QELS), Baltimore, MD, May 18-23, 1997.

Invited conference talk, Ultranarrow Lines in Rare Earth Crystals and Frequency References Based on Persistent Spectral Hole Burning, R. L. Cone, Texas A&M Quantum Optics Workshop on Lasing Without Inversion, Taos, NM, August 14-16, 1997.

Montana State University "OpTeC" Conference on Optical Science and Laser Technology, Bozeman, MT, August 25-26, 1997.

**Invited talk,** Material Design and Characterization for Hole Burning Devices and Optical Frequency Standards, R. L. Cone.

Contributed talk, Erbium based materials for data routing and processing at 1550 nm, Y. Sun, T. L. Harris, R.L. Cone, R.M. Macfarlane, and R.W. Equall.

Contributed talk, Demonstration of Address Header Decoding by Stimulated Photon Echo for Data Routing at the 1550 nm Communications Band in Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>, T. L. Harris, Y. Sun, R.L. Cone, R.M. Macfarlane, and R.W. Equall.

Contributed talk, Persistent Spectral Hole Burning in Rare Earth Doped Deuterated Calcium Fluoride, N. Strickland, R. L. Cone, R. M. Macfarlane.

#### Consultative and Advisory Functions

Cone's group served as advisors on crystal design and characterization to Scientific Materials Corporation, an AFOSR SBIR Phase II contractor. They also worked to enhance linkage of Scientific Materials to other groups in the Spectral Hole Burning community.

Advice was provided to Dr. W. Randall Babbitt, AFOSR contractor, on sensitivity of optical Rabi frequencies, optical nutation, and optical pulse areas to orientationally inequivalent sites in materials with complex crystal structures.

Cone's group built and provided a computer data acquisition system and other instrumentation to AFOSR subawardee R. M. Macfarlane at IBM Almaden Research Center.

A proposal on optical routers for fiber optical systems with terabit bandwidths, more than two orders of magnitude beyond the current state of the art, was prepared and submitted by W. R. Babbitt of the University of Washington and the Cone group to the ARPA Ultra-Photonics program in June, 1996.

Dr. Ralph Kelley of the Atomic Physics Section of AFOSR invited Cone to participate in a Workshop on Time and Frequency Standards on April 25. A talk entitled Spectral Hole Burning Frequency Standards and Stable Lasers was presented at this meeting based on our narrow line spectroscopy, our goals for gated materials and John Carlsten's work on laser frequency locking. The other participants were Selim Shahriar and Philip Hemmer of Hanscom Air Force Base; Ronald Walsworth, Smithsonian Astrophysical Observatory - Harvard, Daniel Kleppner, Physics Department, MIT; Gerald Gabrielse, Physics Department Lyman Laboratory - Harvard; Steve Chu, Physics Department, Stanford; and J.L. Hall, JILA-Boulder.

Professor Marlan Scully invited Cone to participate in the Texas A&M Workshop on Quantum Optics, as there is considerable interest in rare earth materials for new quantum optical phenomena such as lasing without inversion, electromagnetically induced transparency, and quantum computing. On the basis of that talk, a further invitation has been made to the Snowmass Conference, Jan. 4-6, 1998.

Dr. Aleksander Rebane of ETH-Zurich visited our group for proposal preparation with Cone in February, 1997, and joined the Montana State University Physics Department in April, 1997. Rebane's laboratory and office are adjacent to ours, and there will be a strong interaction.

Dr. W. R. Babbitt of the University of Washington visited in May, 1997, and presented a Physics Colloquium. Babbitt joined the Montana State University Physics Department in August, 1997, following a competitive search.

W. R. Babbitt of the University of Washington, Michael Jefferson of IBM – Almaden Research Center, Al Harvey of NSF, Anshel Ghorokhovsky of CCNY, Jean-Pierre Galaup and Jean-Louis Legouet of Paris, and Stefan Kroll of the U. of Lund, Sweden, toured Rufus Cone's research laboratory in conjunction with the AFOSR Workshop on Applications of Persistent Spectral Hole Burning, February, 1997.

Dr. Stefan Kroll of the U. of Lund, Sweden, and Cone's group have agreed to collaborate on dephasing studies.

Dr. Richard Moncorge from Universite Lyon - I and Universite Caen visited August 19 - 26, 1997, to discuss hole burning in rare earth doped materials and laser development.

#### **Transitions**

Development and evaluation of new materials is a special role that we assumed in the AFOSR Photonic Devices and Systems Program of Dr. Alan E. Craig. With Scientific Materials we actively participated in the design of new materials. We cooperatively optimized the material properties through the close interaction with Scientific Materials made possible by our location; by providing rapid feedback. Adjustments were made to the synthesis process, leading to development of better quality materials. Characterized samples of all of the practical materials developed under this AFOSR research grant have become products for Scientific Materials Corporation. Scientific Materials provide research samples to all members of the AFOSR Spectral Hole Burning Applications community at nominal cost.

Materials developed and characterized by Cone's group have been and are being used by a number of other groups in the Spectral Hole Burning Applications community to develop demonstration optical memory and signal processing devices.

Crystals of Eu<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub> grown by Scientific Materials, with our advice and characterization, were used by the Kachru group at SRI to demonstrate optical storage at 10<sup>-8</sup> bit-error rates.

Cone has discussed a material development program with a Los Alamos Group that is working on laser-induced cooling, a technique that may provide the means for practical operation of optical memory or processing devices based on PSHB at lower temperatures.

Crystals of Er<sup>3+</sup>:Y<sub>2</sub>SiO<sub>5</sub>, Er<sup>3+</sup>:CaWO<sub>4</sub>, and Er<sup>3+</sup>:SrWO<sub>4</sub> grown by Scientific Materials, with our advice and characterization, are currently being used by several groups to demonstrate data routing and storage devices at 1.5 µm.

#### New Discoveries, Inventions, or Patent Disclosures

#### New Discoveries, Inventions, or Patent Disclosures

To enable the technical progress on materials for use with semiconductor lasers near 800 nm and 1.5 microns reported above, we identified and arranged production of a variety of Tm<sup>3+</sup> and Er<sup>3+</sup> materials that had not been previously studied by other groups.

A provisional patent application has been processed. The title and abstract are:

# Coherent Interaction of Optical Radiation Beams with Optical-Electronic Materials of Generalized Crystal Symmetry

A method for optimizing the interaction and propagation of a beam (or beams) of radiation through a material having generalized crystal symmetry includes determining a special direction relative to the axes of crystal symmetry of the material and polarizing the radiation beam along this direction. The polarized radiation beam is propagated through the material perpendicular to this special direction.

#### **Honors and Awards**

Principal Investigator Rufus L. Cone received the Wiley Award for Meritorious Research from the Montana State University Foundation, May, 1995.

Randy Wayne Equall completed his Ph. D. in May, 1995. He received job offers from Intel Corporate Research Laboratory and Scientific Materials Corporation. He accepted a permanent position at Scientific Materials Corporation — an AFOSR SBIR Phase II Contractor.

Charles Thiel, an undergraduate working in the group, was a 1995 Congressional Barry M. Goldwater Scholar.

Principal Investigator Rufus L. Cone received the Cox Family Award for Creative Scholarship and Teaching from the Montana State University Foundation, May 10, 1996. In consecutive years Cone thus received MSU's top two academic and research awards.

The Graduate Achievement Award at Montana State University was presented to Randy Wayne Equall for his Ph.D. thesis research on May 10, 1996.

During Autumn Semester, 1995, Gregg White was supported by a Montana Space Grant Fellowship.

The Dean's Award for Excellence in the College of Letters and Science was presented in May, 1996, to Charles Thiel, an undergraduate physics major working in the group. Charles continued his gated spectral hole burning research during Summer, 1996. He entered our graduate program in Physics in August, 1996, and has remained an active member of this research group. He has won both SPIE and Montana Space Grant fellowships.

Tyler Morgus, an undergraduate physics major working in this group, accepted graduate admission to the Ph.D. program at Lehigh University and worked in a research laboratory there during summer, 1996.

Dr. Flurin Koenz of the University of Berne, Switzerland, won a postdoctoral fellowship from the Swiss National Science Foundation for research in Cone's laboratory, October 1, 1996 to September 30, 1997.